

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: Ronald GREINKE

Serial No.: Not yet assigned

Group: 1741

Filed: December 4, 2001

Examiner: Edna WONG

Atty. Docket No.: N-7980

Customer Number: 23456

For: EXPANDABLE GRAPHITE AND METHOD

**PRELIMINARY AMENDMENT REMARKS**

Assistant Commissioner for Patents  
Washington, D.C. 20231

December 4, 2001

Dear Sir:

Further to the continuation patent application filed concurrently herewith, the following amendment remarks are respectfully submitted in connection with the above-identified application.

## **AMENDMENT**

### In the Specification

On page 1, after the title, please insert

#### **-- Priority Information**

This application is a continuation of, and claims priority under 35 U.S.C. § 120 to U.S. Application Serial Number 09/641,835, filed August 18, 2000, now allowed. The entire contents of the parent application are expressly incorporated herein by reference. --

### In the Claims

Please cancel Claims 1-20 without prejudice or disclaimer of the subject matter thereof.

Please add the following claims:

21. A method for preparing expandable graphite flake exhibiting one or more improved exfoliation characteristics, which comprises:
- (a) contacting graphite flake with an organic expansion aid;
  - (b) subjecting said graphite flake to an electrolytic oxidation treatment with an aqueous intercalant solution comprising 10-75% sulfuric acid to provide intercalated graphite flake; and

(c) recovering said intercalated graphite flake.

22. A method according to claim 21 wherein the graphite flake is contacted with said expansion aid prior to subjecting said graphite flake to electrolytic oxidation.

23. A method according to claim 21 wherein the graphite flake is contacted with said expansion aid by dissolving said expansion aid in said aqueous intercalant solution prior to subjecting said graphite flake to electrolytic oxidation therein.

24. A method according to claim 21 wherein said expansion aid comprises a carboxylic acid soluble in said aqueous intercalant solution in an amount effective to enhance exfoliation.

25. A method according to claim 24 wherein said carboxylic acid comprises a carboxylic acid selected from the group consisting of lower aliphatic carboxylic acids and dicarboxylic acids and mixtures of these.

26. A method according to claim 25 wherein said acid comprises a carboxylic acid of the formula  $H(CH_2)_nCOOH$  wherein  $n$  is a number of from 0 to about 5.

27. A method according to claim 21 wherein said intercalant solution contains from about 30 to about 85% water by weight of the solution.

28. A method according to claim 27 wherein said intercalant solution contains from about 50 to about 75% water by weight of the solution.

29. A method according to claim 21 wherein the electrolytic oxidation treatment comprises passing a current between a cathode and the graphite flake as an anode at an anode current density of from about 0.02 to about 0.06 amps per square centimeter.

30. A method according to claim 21, wherein said expansion aid comprises an amount effective to enhance exfoliation of from about 1 to 10% of a carboxylic acid soluble in said aqueous intercalant solution, and said intercalant solution contains from about 30 to about 85% water, all percentages based on the weight of the solution.

31. A method according to claim 21 wherein the electrolytic oxidation treatment comprises passing a current between a cathode and the intercalant wet graphite flake as an anode at a cell voltage of from about 1 to about 6 volts.

32. A method for preparing expandable graphite flake exhibiting one or more improved exfoliation characteristics, which comprises:

(a) contacting graphite flake with an organic expansion aid comprising a carboxylic acid selected from the group consisting of lower aliphatic carboxylic acids and dicarboxylic acids and mixtures of these, said contacting being prior to subjecting said graphite flake to electrolytic treatment;

(b) then, subjecting said graphite flake to an electrolytic oxidation treatment using an aqueous, intercalant solution comprising about 10-75% sulfuric acid to provide intercalated graphite flake, by passing a current through the solution between a cathode and graphite flake wet with the intercalant as an anode at a current density of from about 0.02 to about 0.06 amps per square centimeter and at a cell voltage of from about 1 to about 6 volts; and

(c) recovering said intercalated graphite flake.

33. A method according to claim 32, wherein said expansion aid comprises a carboxylic acid soluble in said aqueous intercalant solution and is employed in an amount of from about 1 to about 10%, both percentages based in the weight of the intercalant solution.

34. An intercalated graphite flake,  
which is contacted with an organic expansion aid, and  
which is subjected to electrolytical oxidization with an aqueous  
intercalant solution that comprises 10-75% sulfuric acid.
35. The intercalated graphite flake of claim 34, wherein  
said expansion aid comprises a carboxylic acid soluble in said  
aqueous intercalant solution in an amount effective to enhance  
exfoliation.
36. The intercalated graphite flake of claim 34, wherein  
said expansion aid comprises an amount effective to enhance  
exfoliation of from about 1 to 10% of a carboxylic acid soluble in said  
aqueous intercalant solution, and said intercalant solution contains from  
about 30 to about 85% water, all percentages based on the weight of the  
solution.
37. The intercalated graphite flake of claim 34, wherein  
said flake is contacted with an organic expansion aid comprising a  
carboxylic acid selected from the group consisting of lower aliphatic  
carboxylic acids and dicarboxylic acids and mixtures of these, said

contacting being prior to subjecting said graphite flake to electrolytic treatment; and then

said flake is subjected to electrolytic oxidation with an aqueous, intercalant solution comprising about 10-75% sulfuric acid by passing a current through the solution between a cathode and graphite flake wet with the intercalant as an anode at a current density of from about 0.02 to about 0.06 amps per square centimeter and at a cell voltage of from about 1 to about 6 volts.

38. The intercalated graphite flake of claim 34, wherein

said flake is subjected to electrolytic oxidation by passing a current through the solution between a cathode and said graphite flake as an anode at a current density of from about 0.02 to about 0.06 amps per square centimeter and at a cell voltage of from about 1 to about 6 volts with said intercalant solution and an organic expansion aid in an amount of from about 1 to 10%.

39. The intercalated graphite flake of claim 34, wherein

said graphite flake is contacted with an organic expansion aid with an intumescent temperature of below about 200°C.

40. The intercalated graphite flake of claim 34, wherein

said graphite flake is subjected to electrolytical oxidization with an aqueous intercalant solution that comprises 10-75% sulfuric acid and from about 30 to about 85% water by weight of the solution.

## **REMARKS**

### Status of the Claims

Claims 1-20 are cancelled. Claims 21-40 are added.

### The Present Invention

The present invention is directed to a process for preparing an intercalated graphite flake having excellent exfoliation characteristics. Furthermore, the present invention is directed to an intercalated graphite flake that is produced by the process of the present invention.

### The Parent Application

This application is a continuation of, and claims priority to, U.S. Application Serial Number 09/641,835, now allowed. In the parent application, a rejection was mailed by the Office which raised several issues with respect to 35 U.S.C. §§ 102, 103. While some of these issues were addressed in the 37 C.F.R. § 1.111 Amendment in the parent



application, to expedite allowance of the parent application allowed subject matter was amended into rejected claims.

In an effort to expedite the prosecution of the instant application, the remaining issues with respect to the parent application are addressed below for the consideration of the Examiner.

#### Issues under 35 U.S.C. § 102

Claims 16-19 of the parent application, corresponding to new claims 34-40 of the instant application were rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Yoshida et al. Reconsideration of this position with respect to this application is respectfully requested.

In order to anticipate a claim, the reference must teach each and every element of the claim. See MPEP § 2131.

Yoshida et al. fail to disclose or suggest an intercalated graphite flake which is subjected to electrolytic oxidation with aqueous intercalant solution that comprises 10 to 75% of sulfuric acid.

The intercalated stage one flakes of Yoshida et al., produced by the chemical reaction with concentrated sulfuric and nitric acid, can be differentiated from the flakes of the present invention by an analysis of the intercalant present in the galleries of the intercalated flakes. The Yoshida et al. intercalated flakes will contain much higher concentrations of internal sulfuric acid (less water) since concentrated

sulfuric acid was experimentally employed. The electrochemical intercalations utilized in the present invention have lower concentration of sulfuric acid.

The Examiner states in the Office Action that "the patentability of a product by process claim does not depend on its method of production and where the Examiner has found a similar product, the burden rests with the Applicant to prove that the product is patently distinct."

However, the claims as written in the present application should not be solely considered as product-by-process claims.

Some apparent process limitations, such as "etched," "welded," "interbonded by interfusion," are considered structural limitations not subject to the product-by-process rules. See In re Garnero, 162 U.S.P.Q. 221, 223 (CCPA 1969).

Additionally, "chemically engraved," read in context, was held to have described the product more by its structure than the process used to obtain it. See Hazani v. United States Int'l Trade Comm'n., 44 U.S.P.Q.2d 1358, 1363 (Fed. Cir. 1997).

Thus, "subjected to electrolytical oxidation..." like "interbonded by interfusion" may be considered as structural limitations not subject to the product-by-process rules. Also, the Examiner should note that these claims (new claims 34-40) have been rewritten to more clearly recite these features.

Claims 16-19 of the parent application were also rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Zhang et al. Reconsideration of this position with respect to this application is respectfully requested.

The intercalated stage one flakes of Zhang et al., produced by chemical reaction with iron chloride in propylene carbonate can be differentiated from the present invention by an analysis of the intercalant present in the galleries of the intercalated flakes. The Zhang et al. intercalated flakes would contain iron chloride and propylene carbonate, and fail to contain the claimed concentrations of sulfuric acid.

Claims 16-19 of the parent application were also rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Kang et al. Reconsideration of this position with respect to this application is respectfully requested.

The intercalated stage one flakes of Kang et al. produced by electrochemical intercalation with pure formic acid can be differentiated from the flakes of the present invention by an analysis of the intercalant present in the galleries of the intercalated flakes. The Kang et al. intercalated flakes would contain formic acid and fail to contain the claimed concentrations of sulfuric acid.

Claim 20 of the parent application was rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Yoshida et al. Reconsideration of this position with respect to this application is respectfully requested.

Yoshida et al. fail to disclose or suggest an intercalated flake with the characteristics described in new claim 34. The Office Action states that "Yoshida teaches an intercalated graphite flake characterized by an intumescent temperature of below about 200° C." Please note that, in addition to the deficiencies with respect to new claim 34, Applicant's review of Yoshida et al. revealed no reference to an organic expansion aid with an intumescent temperature below about 200° C in either the experimental section or in Table I, as indicated by the Examiner.

Claim 20 of the parent application was also rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Zhang et al. Reconsideration of this position with respect to this application is respectfully requested.

Zhang et al. fail to disclose or suggest an intercalated flake with the characteristics described in claim 14. The Office Action states that "Zhang teaches an intercalated graphite flake characterized by an intumescent temperature of below about 200° C." Please note that, in addition to the deficiencies with respect to new claim 34, Applicant's review of Zhang revealed no reference to organic expansion aids with an

intumescent temperature below about 200° C in either the experimental section or in the text of the reference as indicated by the Examiner.

Claim 20 of the parent application was also rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Kang et al. Reconsideration of this position with respect to this application is respectfully requested.

Kang et al. fail to disclose or suggest an intercalated flake with the characteristics described in new claim 34. The Office Action states that “Kang teaches an intercalated graphite flake characterized by an intumescent temperature of below about 200° C.” Please note that, in addition to the deficiencies with respect to claim 14, Applicant’s review of Zhang revealed no reference to organic expansion aids with an intumescent temperature below about 200° C in either the experimental section or in the text of the reference as indicated by the Examiner.

Claims 1 and 3 of the parent application were rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Zhang et al. Reconsideration of this position with respect to this application is respectfully requested.

The Office Action states that the Zhang et al. reference discloses contacting a graphite flake with an organic expansion aid.

However, the Zhang reference discloses propylene carbonate as an expansion aid. Propylene carbonate is not considered an organic

carboxylic acid expansion aid and is inferior to the expansion aids of the present claims. Additionally, the Office Action states that this reference discloses subjecting the graphite flake to an electrolytic oxidation treatment with an aqueous intercalant solution (electrolyte solutions of  $\text{FeCl}_3$ ). However, a solution of  $\text{FeCl}_3$  and propylene carbonate is not an aqueous solution, but rather a nonaqueous solution. In view of such deficiencies, this reference cannot be considered to be anticipatory.

Finally, claims 1, 3, 5-7, and 12 of the parent application were rejected under 35 U.S.C. § 102(b) as allegedly being anticipated by Kang et al. Reconsideration of this position with respect to the present application is respectfully requested.

Kang et al. fail to disclose a graphite flake electrolytic oxidation treatment with an aqueous intercalant solution comprising 10-75% sulfuric acid. In view of this deficiency, this reference cannot be considered to be anticipatory.

#### Issues under 35 U.S.C. § 103

Claims 2, 8-9, and 11-13 of the parent application were rejected under 35 U.S.C. § 103 as allegedly being obvious over Zhang et al. Reconsideration of this position with respect to this Application is requested. The deficiencies of the Zhang et al. reference with respect to the present invention are discussed above.

In order to establish a *prima facie* case of obviousness, there must be some suggestion or motivation to modify the reference to teach or suggest all of the claim limitations with a reasonable expectation of success. See MPEP §2143. In view of the differences stated above, which at least do not suggest all of the claim limitations, a *prima facie* case of obviousness cannot be established with respect to this claim and this rejection.

Claims 2 and 11 of the parent application were rejected under 35 U.S.C. § 103 as allegedly being obvious over Kang et al. Reconsideration of this position with respect to this Application is requested.

In view of the differences stated above, which at least do not suggest all of the claim limitations, a *prima facie* case of obviousness cannot be established with respect to this claim and this rejection.

Finally, claim 13 of the parent application was rejected under 35 U.S.C. § 103 as allegedly being obvious over Kang et al. Reconsideration of this position with respect to this Application is requested.

In view of the differences stated above in connection with this reference, a *prima facie* case of obviousness cannot be established with respect to this claim and this rejection.

Conclusion

From the foregoing, further and favorable reconsideration in the form of a Notice of Allowability is on order, and such action is earnestly solicited.

In an effort to expedite the prosecution of this Application, if there are any issues precluding immediate allowance, or if the Examiner has any questions regarding these Remarks or the Application in general, she is respectfully urged to contact Richard S. Myers, Jr. (Reg. No. 42,022) at the number listed below.

If necessary, the Commissioner is hereby authorized in this, concurrent, and further replies, to charge payment or credit any overpayment to Deposit Account No. 50-1202 for any additional fee required under 37 C.F.R. 1.16 or under 37 C.F.R. 1.17; particularly, extension of time fees.

Respectfully submitted,



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